Precise Investigation of FISDW in π-d Hybrid Molecular Conductors

M. Maesato (Kyoto U., Chemistry); G. Kawaguchi (Kyoto U., Chemistry); H. Kitagawa (Kyoto U., Chemistry); T. Imakubo (Nagaoka U. of Tech., Materials Science and Technology); A. Kiswandhi (FSU, Physics); D. Graf (NHMFL); J.S. Brooks (FSU, Physics)

Introduction

The quasi–one dimensional (Q1D) molecular conductors $(DIETSe)_2FeX_4$ [X=Cl, Br] are multifunctional π–d hybrid materials having a spin density wave (SDW) instability of π-electrons and an antiferromagnetic (AF) order of d-electron spins, where DIETSe denotes diiodo(ethylenedithio)tetraselenafulvalene [1]. The $FeCl_4$ salt undergoes a metal-SDW transition at 12 K, followed by an AF transition of d-electron spins at 2.5 K. Therefore, SDW and AF order coexist in the ground state. On the other hand, the isostructural $FeBr_4$ salt shows an AF transition at 7 K, associated with a metal-insulator transition. It suggests an AF-order induced SDW transition in the $FeBr_4$ salt. We have reported spin-flop induced sharp positive magnetoresistance as large as 100 % and nonvolatile magnetoresistive memory in $(DIETSe)_2FeCl_4$ salt, indicating the interplay between the itinerant π-electrons and local moments of d-electron spins[2]. The application of high pressure suppresses the SDW phase, due to the increase in the Fermi surface warping[2,3]. Recently, we have succeeded to discover field-induced SDW (FISDW) states in $(DIETSe)_2FeCl_4$ under high pressures. We also observed quite anomalous magnetoresistance in the isostructural $(DIETSe)_2FeBr_4$ salt. It showed very large hysteresis in the magnetoresistance below 24 T at 0.3 K [4]. The magnetic torque measurements revealed that the critical field corresponds to the boundary of AF transition of d-electron spins.

In order to systematically control the electronic states and π–d interaction of DIETSe salts, we have prepared a series of mixed crystals $(DIETSe)_2FeCl_{4x}Br_{4(1-x)}$ and investigated high magnetic field effects on transport and magnetic properties.

Experimental

The magnetoresistance was measured under hydrostatic pressure up to 35 T using the DC magnet in Cell12. Magnetic field was applied along the least conducting *b*-axis direction. We used clamp-type small BeCu pressure cells and Daphne oil 7373 as the medium for the high pressure application. Pressures were calibrated by monitoring the resistance change of manganin wire. Magnetic torque was measured utilizing a cantilever method.

Results and Discussion

We have found anomalous behavior in the magnetoresistance of $(DIETSe)_2FeCl_{4x}Br_{4(1-x)}$ (x=0.5, 0.75), associated with large hysteresis. A hysteresis also appeared in the magnetic torque at the same magnetic field, indicating the strong correlation between mobile electrons and local moments. The critical field corresponds to the boundary of the AF transition, which increases with increasing x, indicating the increase in the π–d interaction. We also observed quantum oscillations in the magnetoresistance, confirming the high quality of the mixed crystal we have studied. The oscillations are attributable to the existence of a closed orbit or the quantum interference effect. In addition, a large increase in magnetoresistance was observed at low temperatures and strong magnetic fields, reminiscent of FISDW transition. Such behavior was suppressed with increasing x, suggesting an increase in the Fermi surface warping. Therefore, we have successfully controlled, in a systematic way, the electronic states and π–d interactions of mixed crystals $(DIETSe)_2FeCl_{4x}Br_{4(1-x)}$.

Acknowledgements

This work was supported by KAKENHI (23225005) by JSPS, NSF-DMR-0654118, the State of Florida, and the DOE, and by NSF-DMR-1005293 (JSB).

References

[1] Shirahata, T, *et al.*, J. Mater. Chem., **2006**, 16, 3381-3390 (2006).
[2] Maesato. M., *et al.*, J. Am. Chem. Soc. **134**, 17452 (2012).
[3] Maesato. M., *et al.*, submitted to Phys. Rev. B.
[4] Maesato. M., *et al.*, submitted to J. Phys. Soc. Jpn.